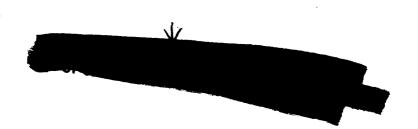
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DETONATION WAVES IN GASES

R. I. Soloukhin

TABLE OF CONTENTS

1.	Introduc	tion	1
2.	Structur	e of the Detonation Wave	1
	2.1.	The Steady-State Zone of the Chemical Reaction	2
	2.2.	The Rarefaction Wave	5
	2.3.	The Chapman-Jouguet Condition	7
المارين المارين	2.4.	Divergent and Convergent Detonation Waves	9
3.		ntal Methods of Studying the State of the ehind the Detonation Front	10
	3.1.	Pressure Measurements (Pulsed Piezoelectric Pressure Gauges)	11
. 3	3.2.	Gas Density Measurements	13
	3.3.	Gas Temperature Measurements (Generalized Method of Rotation of Spectral Lines)	15
	3.4.	Determination of the Chemical Composition and Concentrations of Individual Components of the Mixture	16
4.	Characte	ristics of Gas Ignition Behind the Shock Front	17
	4.1.	The Induction Period	18
	4.2.	Determination of the Kinetic Reaction Parameters	22
	4.3.	Development of the Detonation Front	21

5•	Transiti	on from Deflagration to Detonation in Gases	24
	5.1.	Gasdynamic Scheme of the Development of Detonation in a Tube	25
	5.2.	Calculation of the State of the Mixture in Front of the Flame Front	26
	5.3.	The Compression Wave and Adiabatic Self-Ignition .	28
	5.4.	Interaction of Flame and Shock Wave	31
6.	Oscillat	ion of the Gas Behind the Detonation Front	32
	6.1.	Causes of the Appearance of the Oscillating Combustion Regime	33
	6.2.	Properties of Transverse Waves	34
	6.3.	Acoustic Theory of Spinning Detonation	35
	6.4.	Structure of the Spinning-Detonation Front	37
	6.5.	State of the Gas in the Detonation Wave Allowing for Transverse Pulsations	3 8
7.	Detonati	on in a Steady Gas Flow	40
	7.1.	Combustion of Gas Behind a Steady Shock Wave	4C
	7.2.	Steady-State Spinning Detonation	40
	7•3•	Pulsating Combustion Behind a Shock Wave in a Supersonic Flow	42
8.	Conclusi	ons	43
Bib	liography	·	إلبإ

1. INTRODUCTION

Detonation waves represent a complex of interrelated physico-chemical processes; hence, aside from its technical significance, the problem of detonation has attracted the attention of many researchers in various branches of knowledge. The establishment of thermodynamic equilibrium behind the detonation front, oscillations of the gas and spinning detonation, the transition from slow combustion to detonation, detonation limits, combustion and detonation in supersonic gas flows --all these problems have proved to be closely related to the characteristics of shock waves, to the rates and mechanism of chemical reactions, turbulence, acoustic vibrations, and other processes in gases. The study of detonation has not been limited exclusively to the classical Chapman-Jouguet scheme. Considerable deviations from this scheme, relating to the characteristics of the physicochemical and hydrodynamic processes in a gas medium, were noted long ago.

The investigation of detonation phenomena has made it necessary to develop very precise methods of making pulsed microsecond measurements of various quantities: pressure, density, temperature, the emissivity and absorptivity of gases in various spectral regions, chemical composition, and concentration. Improving shock-tube measuring techniques [1-4] have made it possible to use many of the methods developed to study the dynamics of ignition and to determine the state of the gas behind the detonation front.

The most interesting results of investigations of high-speed processes in detonation waves are discussed below. The foreign surveys of the problem [5-7] are far from complete, especially with respect to the work of Soviet authors. Moreover, this approach is not always adequately represented in the programs of international symposia on combustion and detonation. For example, the problem of spinning detonation, to the solution of which many of the investigations of recent years have been devoted, is clearly receiving insufficient attention. Equally neglected is the problem of the kinetics of chemical reactions at temperatures corresponding to detonation conditions.

2. STRUCTURE OF THE DETONATION WAVE

Detonation waves are a special variety of discontinuous gasdynamic flow. In a medium capable of an exothermic reaction, a selfsustaining shock discontinuity is formed. Behind this shock front a chemical reaction is continuously initiated as a result of heating due to adiabatic compression. The detonation speed is related to the

/526

velocity of the shock wave and comprises several kilometers per second, which is hundreds of times greater than the flame velocity in gases, as determined by the laws of propagation of a thermal front. A detailed analysis of the shock adiabatic curve in detonation and calculations of the velocity of detonation waves in various gases will be found, for example, in [7-9]. Here we shall discuss only certain details of gas flow in the detonation wave.

2.1. The Steady-State Zone of the Chemical Reaction

The physical model of the detonation wave still remains unclear in many respects. This applies particularly to the structure of the chemical reaction zone. The classical one-dimensional scheme proposed by Chapman and Jouguet is the simplest. According to this scheme, the chemical reaction proceeds instantaneously in the shock wave, while expansion of the burned products occurs in the isentropic rarefaction wave that follows the detonation front.

In spite of the fact that in many cases it is sufficient to limit oneself to this assumption in order to find the detonation speed, a number of the characteristics of detonation waves are related precisely to the final value of the chemical reaction rate. For example, if the friction losses and heat losses in the reaction zone exceed the supply of energy from the chemical reaction, damping of the detonation wave, known as the phenomenon of the "detonation limit," sets in. The problem of detonation with losses is analyzed in detail in [8] on the basis of the hypothesis of a steady-state reaction zone.

As will be seen from the following, the existence of an extended chemical reaction zone leads to considerable deviations from a steady-state and one-dimensional gas flow behind the detonation front. None-theless, the one-dimensional model of the detonation wave with a steady-state chemical reaction zone behind the shock front (Ya. B. Zel'dovich [10, 8], subsequently [11-13]) is a considerable refinement of the scheme of the phenomenon and in a number of cases correctly describes the structure of the gas flow.

The sequence of the processes developed in the detonation wave is illustrated by Fig. 1, which schematically shows the profile of the change in gas temperature and pressure. After the shock transition (1-2), which is completed in the course of several molecular collisions, rotational and vibrational relaxation occurs -- energy exchange among the translational, rotational, and vibrational degrees of freedom of the gas molecules. In the relaxation zone the gas temperature corresponding to the energy of translational motion drops, while the pressure and density of the gas increase to the equilibrium values.

Thanks to the use of methods utilizing shock waves, the relaxation processes in gases at high temperatures have been sufficiently well studied [1, 14]. Although the excitation of molecular rotations is completed in several dozen collisions, the vibrational relaxation

continues for 10^4 - 10^6 collisions. At atmospheric pressure and a temperature of 1500-3000°K, the vibrational relaxation time for oxygen, for example, is from 1.5 to 15 microseconds.

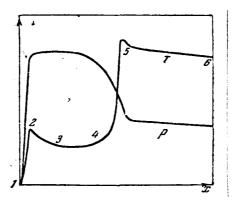
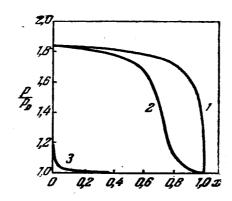


Fig. 1. Variation of pressure and temperature behind the detonation front (no scale).

The relaxation processes of excitation of vibrations, as well as possible dissociation of the starting materials, precede the chemical reaction, which is completed in 10^{12} - 10^{14} collisions. The reaction zone (3-5) has its own characteristics. For chemical reactions with a sufficiently high activation energy (E = 20-40 kcal/mole), the latent, induction period of the reaction may comprise more than 90% of the entire reaction time [15]. During the induction period, active reaction centers accumulate, while the pressure, density, and temperature of the mixture vary little. The induction stage of the reaction (3-4) ends in explosion (4-5). The ratio of the induction and explosion periods is determined by the reaction mechanism. Fig. 2 shows the results of calculating the pressure change behind the front of a one-dimensional steady-state detonation in a mixture of $2H_2 + 0_2$ for various

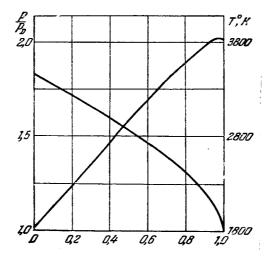
cases of dependence of the reaction rate on the concentration of the



[commas represent decimal points]

Fig. 2. Profile of pressure in reaction zone.

1 - zero-order reaction; 2 - first-order reaction; 3 - second-order reaction (according to [16]).



concentration ratio

[commas represent decimal points]

Fig. 3. Pressure and temperature in the reaction zone as a function of the ratio of the fluid concentration of the final products to their equilibrium value [16].

products obtained (zero-, first-, and second-order reactions) [16]. The calculated dependence of the pressure and temperature in the intermediate state of the reaction zone on the relative concentration of the reaction products in the same mixture is shown in Fig. 3.

In an established detonation wave, the profile (1-5) is assumed to be steady-state. The structure of the reaction zone is determined by simultaneous solution of the hydrodynamic equations of the conservation of mass, momentum, and energy in steady flow and the equations of chemical kinetics. We must mention at once, however, that usually in self-sustaining detonation waves a one-dimensional steadystate reaction zone is not realized. An increasing number of investigations, of which we shall speak later, gives convincing evidence that the gas flow in the reaction zone is far from one-dimensional and is accompanied by intensive transverse disturbances. Especially noteworthy are the latest data of D. Hornig [17], in which deviations from unidimensionality have been detected even in detonation waves in hydrogen-oxygen mixtures at high initial pressures (up to 20 atm). Moreover, significant deviations from unidimensionality are observed not only in gases, but also in the detonation of liquid explosives [18]. The reaction zone is bounded by a region of nonsteady flow -the rarefaction wave (5-6), the profile of which varies with time in accordance with the boundary conditions for the flow of combustion products.

2.2. The Rarefaction Wave

The flow of burned gas in a one-dimensional rarefaction wave is described by the following relations:

$$x = (u + a) t$$

and

$$u_2 - u = \frac{2}{\gamma - 1} (a_2 - a),$$

where x denotes the coordinate of the gas particle, u the flow velocity, and a the speed of sound. The subscript 2 denotes the state of the gas at the boundary between the reaction zone and the rarefaction wave. According to the well-known Chapman-Jouguet condition, the detonation speed $D = u_2 + a_2$ (see paragraph 2.3); hence

<u> _528</u>

$$x_2 = Dt = (a_2 + u_2) t.$$

Consequently,

$$u_2 - u = \frac{2}{\gamma + 1} \left(\frac{x_2 - x}{t} \right).$$

If we neglect the gas pressure in front of the detonation front as compared with the pressure behind it: $p_2 \gg p_1$, from the laws of conservation we can obtain:

$$u_2 = D - a_2 \cong \frac{a_2}{\gamma}, \quad u_2 - u = \frac{2a_2}{\gamma} (\frac{x_2 - x}{x_2}).$$

Thus, for detonation in a long tube the velocity of the burned gas falls linearly as the distance from the front x increases. When

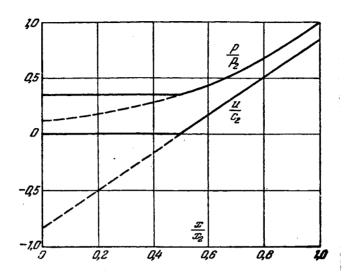
 $x = \frac{x_2}{2}$, u = 0, i.e., starting from this point, in a tube with a closed end, the gas will be at rest with respect to the walls of the tube. If the detonation wave is initiated at the open end of the

tube, at the point $x = \frac{x_2}{2}$ the velocity of the gas will change sign. The distribution of the gas pressure is found analogously:

$$\frac{P}{P_2} = \left(1 - \frac{\gamma - 1}{\gamma} \frac{x_2 - x}{x_2}\right)^{\frac{2\gamma}{\gamma - 1}}.$$

In Fig. 4 the change in pressure and velocity along the detonation tube for the case of initiation at the closed end (x=0) is shown by solid lines. The broken lines correspond to the flow at the open end of the tube. The calculations were made for $\gamma=1.2$, which closely corresponds to the flow of the detonation products of a mixture of hydrogen and oxygen. An experimental verification of the density variation in the rarefaction wave during detonation in a tube may be found in [19].

<u> _529</u>



[commas represent decimal points]

Fig. 4. Flow structure in a one-dimensional detonation wave.

Broken line — tube with open end, x — coordinate of detonation

front,
$$\gamma = c_{p}/c_{v} = 1.2.$$

2.3. The Chapman-Jouguet Condition

The state of the gas determined by the so-called Chapman-Jouguet condition is the boundary between the steady-state reaction zone and the rarefaction wave. From the equations describing the conservation of mass and momentum in a one-dimensional isentropic gas flow, we can obtain the following relation by introducing the variable $\xi = x/t$:

$$\frac{du}{d\xi} \left[1 - \frac{(\xi - u)^2}{a^2} \right] = 0.$$
 (I)

Consequently, the position of the boundary of the nonsteady-state

rarefaction wave $(\frac{du}{d\xi} \neq 0)$ should be determined by the condition

$$\xi_{fr} = D = u + a,$$

i.e., the propagation velocity of the detonation front should be sonic with respect to the particles of burned gas. Thermodynamic considerations also lead to the same function [8, 13].

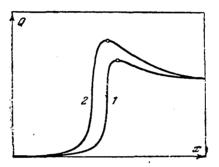


Fig. 5. Displacement of the curve of heat liberation in the reaction zone, caused by inhibition of dissociation and an increase in the detonation speed.

l - variation of heat liberation behind the front of a normal detonation wave with the distance from the shock front; 2 - the same curve with the influence of the detonation speed and pressure taken into account. The Chapman-Jouguet state is close to the point of the maximum.

Certain comments must be made concerning the Chapman-Jouguet condition. This relation serves as a basis for calculating the detonation speed in various gases [9, 20]; hence an accurate determination of the speed of sound in the combustion products is important. However,

the speed of sound is not an entirely unambiguous quantity, owing to the dispersion caused by retardation of the excitation of molecular vibrations. There is every reason to believe that the speed of sound will be greater for the high-frequency than for the low-frequency components. In addition, it will not always be correct to calculate the thermodynamic state of the reaction products from the conditions of chemical equilibrium and the total heat evolution at the Chapman-Jouguet boundary. For example, the rates of certain dissociation reactions of the combustion products may be less than those of the reactions determining the basic evolution of heat. In this case the Chapman-Jouguet boundary is displaced in the direction of the zone of the basic chemical reaction, while slow reactions will no longer exert an influence on the parameters of the steady-state zone of chemical transformations. The inhibition of dissociation and the existence of nonequilibrium concentrations of the final products at the Chapman-Jouguet boundary are equivalent to greater heat liberation in the wave as compared with the equilibrium calculation. As a result, the detonation speed proves to be higher than the equilibrium value, while the gas pressure in the Chapman-Jouguet state also increases. Increasing the pressure will promote the suppression of dissociation and the displacement of the Chapman-Jouguet boundary into the region of higher heat evolution within the limits of the steady-state reaction zone (Fig. 5). Thus, the proper selection of the detonation speed should take into consideration displacement of the boundary of the steady-state reaction zone corresponding to the increase in total heat liberation and pressure in this zone. This interesting feature of the structure of the detonation wave was first noted by Ya. B. Zel'dovich [9, 10]. Certain experimental data also indicate the possibility of the existence of such combustion regimes in the detonation wave [21]. However, as has already been noted, such an effect may be complicated to a considerable degree by deviations from unidimensionality of the gas flow in the vicinity of the detonation front.

2.4. Divergent and Convergent Detonation Waves

By examining three-dimensional detonation waves in the case of spherical or cylindrical symmetry, we can obtain a graphic idea both of the broad region of applicability of the Chapman-Jouguet scheme (plane and divergent waves) and of the existence of physically substantiated deviations from this scheme ("focusing" or convergent waves). For nonplanar three-dimensional waves, equation (I) has a right-hand side, which per se is evidence of the possibility of deviations from steady-state wave propagation. An analysis of the gas flow in divergent

detonation waves [8, 22, 23] has shown that behind the front in a divergent flow only the law of expansion of the gas in the rarefaction wave adjoining the reaction zone changes. In this case steady-state wave motion is preserved, as confirmed by experiments, in which non-damping spherically [24] and cylindrically [25] divergent detonation waves were observed. As a curious fact, we should mention that according to (I) large gradients of the velocity and other parameters of the gas behind the detonation front should exist for a steady-state divergent wave in the Chapman-Jouguet state. This circumstance at one time served as ground for doubting the possibility of the existence of divergent waves without damping (Jouguet).

Although the variation of the law of expansion of the combustion products behind the detonation front exerts no influence on the velocity of the wave, in relation to added compression this is true only within certain limits. By artificially increasing the compression behind the detonation front, for example, by means of a piston inserted into the tube behind the wave, the pressure drop in the rarefaction wave following the reaction zone can be partially or entirely compensated [26]. In this case the steady state and speed of the detonation are maintained. However, if the velocity of the piston is made greater than the flow velocity of the gas particles in the Chapman-Jouguet state, the speed of the detonation front increases, in accordance with the laws of conservation at a shock discontinuity, and will depend on the velocity of the piston. The so-called supercompressed detonation is formed [27].

As Ya. B. Zel'dovich has shown, supercompression sets in spontaneously in a convergent wave on account of the increase in compression in the focusing flow. Utilizing the tendency of a detonation to retain its steady-state characteristics, we can calculate the pressure change in the wave front during the focusing process [28]. In the limiting case, at very small radii of the convergent front, the behavior of the detonation wave will differ little from the focusing of a shock wave, i.e., the chemical energy liberated can be neglected in comparison with the work of compression in the convergent flow. The asymptotic law of the pressure increase in the wave front can be determined from the known solutions given in [29].

3. EXPERIMENTAL METHODS OF STUDYING THE STATE OF THE GAS BEHIND THE DETONATION FRONT

The chemical reaction behind the detonation front is completed within very short intervals of time. For example, at atmospheric pressure the induction period of the reaction behind the shock front in

the detonation of a mixture of hydrogen and oxygen is about 2 microseconds. Consequently, the resolving power of the measurements designed to register the time variation of various physical quantities

in the detonation wave should be at least less than 10⁻⁶ sec. Let us briefly consider some of the methods of making such measurements (a more detailed description may be found in [3]).

3.1. Pressure Measurements (pulsed piezoelectric pressure gauges)

The most accurate and convenient method of measuring the pressure behind the shock-wave front is that of pulsed piezoelectric gauges, utilizing ceramic piezoelectric materials [4, 30-32]. Tourmaline, quartz, Rochelle salt, and other natural piezoelectric materials are less sensitive, less reliable in operation, and more prone to excite parasitic vibrations in the volume of the piezoelectric element.

Essentially, the accuracy of the measurements can be improved and the resolving time of the gauges reduced in two different ways. First, special attention may be paid to reducing the linear dimensions of the piezoelement, while, secondly, an attempt may be made to create conditions corresponding to the propagation of an elastic wave in a piezoelement of finite dimensions, permitting an exact determination of the pressure acting on the sensitive surface.

The construction of a pulsed piezoelectric pressure gauge for measuring the gas pressure at the wall of the tube is shown in Fig. 6. A cylindrical piezoelement made of barium titanate ceramic is soldered to a long zinc rod so as to draw off into the rod, without reflection, the elastic wave, propagated in the piezoelement following a sharp pressure change in the gas. The acoustic resistances of the zinc and barium titanate are similar in magnitude [30]. To eliminate any acoustic coupling between the piezoelement and the walls of the tube, the space between the casing and rod is filled with beeswax or some other "decoupling" filler. Samples of pressure readings made during the shock-tube detonation of the mixtures 2H + 0, 2CO + 0, and a

mixture of ethyl alcohol and oxygen are shown in Fig. 7. In this arrangement the diameter of the piezoelement can be reduced to 1-1.5 mm, while maintaining a sensitivity of \sim 10 v·pf/atm. The resolving time of the gauge may be up to 0.5 μ sec.

In [31] the present writer describes the mode of operation of a pulsed piezoelectric pressure gauge that combines a short resolving time and a large piezoelement. In this case the pressure p(t) acting on the sensitive surface is reproduced in terms of the registered curve

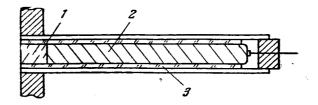


Fig. 6. Pulsed piezoelectric pressure gauge.

1 - piezoelement; 2 - zinc column; 3 - wax
filler.

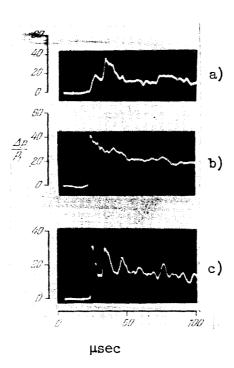


Fig. 7. Oscillograms of the pressure behind a sustained detonation in a shock tube with a square cross section.

- a) mixture of carbon monoxide and oxygen;
- b) mixture of ethyl alcohol vapor and oxygen;
- c) mixture of hydrogen and oxygen. Initial gas pressure 0.1 atm.

q(t) of the increase with time in the charge on the piezoelement as the elastic compression wave is propagated in it:

$$q(t) = A \int_{0}^{t} p(t) dt.$$

This method was used to measure the pressure correct to 0.4 μ sec for normal reflection of a detonation wave from a rigid wall; the existence of a region of increased pressure, corresponding to the chemical reaction zone, was also demonstrated.

3.2. Gas Density Measurements

The Schlieren method [3, 4, 33] and the interferometer method [34-36] are widely used to measure the variation of the gas density behind the shock-wave front. Both methods are based on the registration of the change in the refractive index of the gas and are very sensitive (a change of as little as 0.1% ϱ_1 is registered). Fig. 8

shows an interferogram of the flow structure behind the detonation front in a mixture of $2H_2$ + 0.92 Xe (p_1 = 40 mm Hg). The vertical

displacement of the interference bands is proportional to the change in gas density. The fine details of the flow, registered by the interferometer, may make it difficult to obtain data on the average values of the density close to the detonation front. Hence, the method of absorption of a thin beam of X-rays, directed perpendicular to the motion of the wave front, is considered the most convenient method of measuring density in the study of detonation [19, 21, 37-39]. In this case an inert gas, usually xenon, is added to the investigated mixture to increase the absorptivity of the gas. The variation in radiation intensity is registered by means of a photomultiplier, fitted with a scintillator. Fig. 9 shows a comparison of the calculated and measured profiles of the density of the detonation wave in a mixture of $2H_0$

+ 0.92 Xe at an initial pressure of the mixture of 44 mm Hg. The

observed discrepancy cannot be attributed solely to the limited resolving power of the method of density measurement. The detonation

/533

waves, illustrated in Figs. 8 and 9, were obtained almost under the same conditions [36, 38]. As we can see from Fig. 8, the shock front of the detonation is not a plane surface and is strongly disturbed by transverse waves (see also Section 6.2). Hence, in measuring the density from the absorption of X-rays, not a one-dimensional chemical reaction zone, but some average profile of the density variation over the front is registered.

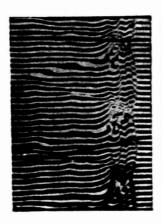


Fig. 8. Interferogram of the propagation of a detonation in a mixture of $2H_2 + 0_2 + 0.92$ Xe.

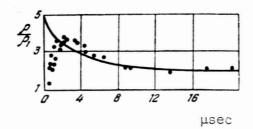


Fig. 9. Results of experiments to measure the density behind the detonation front (X-ray absorption).

In the experiments of D. Hornig [17], the variation of the gas density behind the detonation front was registered by the method of reflection of light from the surface of the front. This method made it possible to determine the structure of the shock front in ordinary gases with high resolution (of the order of the wave length of the light) [40]. Measurements in detonation waves showed that the jump in gas density in the front corresponds to a shock transition without chemical reaction. However, the effective width of the reaction zone proved to be far greater than expected. Thus, for example, in a mixture of $2H_2 + O_2$, at an initial pressure of 20 atm, the width of the transition zone is about 2.5 mm, which is hundreds of times greater than the width of the one-dimensional reaction zone indicated by kinetic data. These results are evidence of considerable deviations from unidimensionality, even at high pressures of the mixture.

3.3. Gas Temperature Measurements (generalized method of rotation of spectral lines)

This method proved to be most convenient for studying the state of the gas behind the shock-wave front [41-44]. Although pyrometric measurements of the emissivity in two portions of the continuous emission spectrum are sufficient to determine the gas temperature in the detonation of condensed explosives [45-57], in the study of detonation in gases, it is necessary to make an independent determination of both the emissivity and the absorption of light in the spectral regions of intense radiation of the gas or the impurities it contains. When the spectral lines of the emission of the impurities are used to measure the gas temperature (see [41, 42], as well as the surveys [1, 48]), the resolving power is limited by the time required for the heating, evaporation, dissociation, and excitation of the impurities (for example, sodium salts) in the gas flow behind the shock wave. The possibility of measurements close to the shock front is thereby excluded. Thus, the lag in establishing thermal equilibrium in the emission of the D line of sodium behind the shock-wave front in air may amount to several dozen microseconds [42].

In [49] an attempt was made to measure the temperature behind the detonation front in an acetylene-oxygen mixture from the rotation of the lines of the intrinsic radiation of the gas. Such measurements are possible thanks to the concentration of the intermediate radical C_{\odot} in the chemical reaction zone. Fig. 10 shows oscillograms of the

luminescence of the gas 1 and luminescence of the gas with bias lighting from a standard light source 2. A pulsed krypton-xenon lamp, the <u> /534</u>

color temperature of which may reach 5000°K, was used in these experiments as a comparison light source. The resolving time of such

measurements is up to 0.5·10⁻⁷ sec. The measured gas temperature for various concentrations of the components of an acetylene-oxygen mixture proves to be close to the equilibrium calculation.

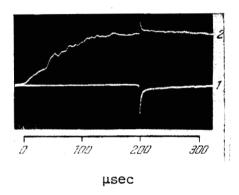


Fig. 10. Oscillograms of the emission of the gas behind the detonation front in a mixture of $C_2H_2 + O_2$, $\lambda = 5165$ Å.

3.4. Determination of the Chemical Composition and Concentrations of Individual Components of the Mixture

Here, too, spectral methods are widely used. For example, the method of measuring the concentration of molecular oxygen from its absorption in the ultraviolet region has been well developed (S. A. Losev and N. A. Generalov [1, 50]). If the dependence of the absorptivity of molecular oxygen on the temperature and concentration is found experimentally, the variation of the absorption behind the shock front will uniquely determine the variation of the absolute oxygen concentration in the mixture. As indicated in [51], a layer of oxygen 1 cm thick absorbs about 30% of the transmitted light in the region $\lambda = 1470$ Å at room temperature and a pressure of 1 mm Hg. The method of absorption spectroscopy in the ultraviolet region has been successfully used to determine the rate of dissociation of pure oxygen

and the oxygen in air in strong shock waves [50].

Analogously to investigations in flames [15, 52], the kinetics of chemical reactions behind the fronts of shock and detonation waves can be studied by observing the variation with time of the absorptivity of certain intermediate reaction products. For example, Bauer et al. [53] have studied the decomposition of water vapor behind a shock-wave front. The measurements were performed at a temperature of $2400-3200^{\circ}$ K in the region of hydroxyl absorption, $\lambda = 3064$ Å.

Nonequilibrium concentrations of carbon monoxide and atomic oxygen were detected in the reaction zone behind the detonation front in a mixture of $2H_2 + O_2 + 2CO$. It was found that the emissivity of

the gas in individual portions of the visible region of the spectrum is proportional to the product of the concentrations (CO) (O) [36]. In these experiments the amplitude of the luminescence in the reaction zone corresponded to the concentration of the reaction products, calculated without considering the slow recombination reactions that occur in the case of triple collisions. The length of the reaction zone registered by this method was several millimeters at an initial gas pressure $p_1=0.1\text{--}0.3$ atm. As the measurements showed, the data

on the parameters of the reaction zone are close to the results of one-dimensional calculations only in "supercompressed" waves, while self-sustaining detonation waves display strong deviations from unidimensionality.

Thus, while we possess sufficiently well developed methods of making physical measurements in detonation waves, we do not as yet have at our disposal reliable data on the state of the gas directly behind the detonation front. The difficulties arising here are not technical, but theoretical in character, since the existing limitations relate primarily to the substantial deviations from unidimensionality of the gas flow behind the front of the detonation wave. The causes of these deviations will be discussed in the following sections.

4. CHARACTERISTICS OF GAS IGNITION BEHIND THE SHOCK FRONT

The thermodynamic state of the gas behind the shock front of a detonation is determined by the propagation velocity and by the initial pressure of the mixture. The gas temperature is usually 1500-2000°K, which corresponds to an ignition delay of about 1 microsecond at pressures of the order of atmospheric. The kinetics of the chemical reactions under these conditions have been little investigated;

hence when the well-known reaction mechanisms are used in the high-temperature range, a special experimental verification of their applicability in this case is required. Shock waves can be effectively used to study the kinetics of ignition in the case of microsecond delays [54-62]. The shock-wave method possesses a number of advantages in comparison with other methods of chemical kinetics, since the thermal effect of the walls of the vessel is essentially excluded. In addition, the temperature range of investigations in shock waves is far broader than usual.

4.1. The Induction Period

The method of reflected waves can be used to study the dependence of the induction period of a mixture on the temperature and pressure. In this case the ignition conditions of the mixture are dictated by the state of the gas behind the wave front in the tube after reflection from the closed end. Fig. 11 shows the time development of the process of ignition in a mixture of $\rm H_2 + \rm O_2$, made by the Schlieren method.

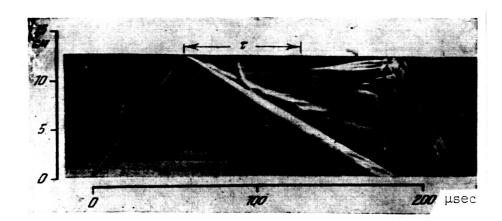


Fig. 11. Development of ignition with delay, made by the Schlieren method in a mixture of $H_2 + O_2$.

The initial mixture is compressed twice -- behind the incident wave 1 and the reflected wave 2. The state of the gas behind the front of these waves is homogeneous, while the gas particles behind the front of wave 2 are at rest with respect to the walls of the tube. The pressure, density, and temperature of the gas are usually determined by gasdynamic calculations. A check of the validity of such calculations by means of independent piezoelectric pressure measurements and density measurements using an interferometer and the Schlieren method did not lead to deviations greater than 1-2%. Thus, for example, Fig. 12 shows the results of measurements of the flow velocity of the gas behind wave 1 in a mixture of $200 + 0_2$, obtained from the slope of the

<u>/536</u>

contact lines on Schlieren photographs of the process [3]. The experimental points usually lie somewhat above the calculated curve, which serves as a measure of the deviation from unidimensionality in shock tubes [3, 63].

A chemical reaction develops within the gas volume heated by the shock wave. In order to avoid taking into account the effect of the heating of the gas in the incident wave 1, the measurement of the induction period is usually related to the layers of gas adjoining the reflecting wall. Inasmuch as the gas ignites simultaneously in several points of the gas volume 3, the values of τ (see Fig. 11) are extrapolated to $\tau_{\rm wall}$ [55]. Focal ignition of the gas, beginning in the form

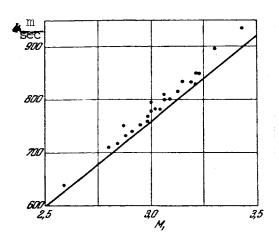
of deflagration from individual centers, culminates in explosion and in the formation of a detonation wave 4.

Fig. 13 presents the results of measurements of the ignition delay in a mixture of $\rm H_2$ + $\rm O_2$ at atmospheric pressure in the tempera-

ture range from 1000 to 1600°K. The reaction kinetics of this mixture at lower pressures and temperatures are sufficiently well known; hence the data obtained can be compared with the delays determined from the chain mechanism of this reaction [15, 64, 65]. During the induction period, when the concentrations of the reaction products are low, the reaction rate is determined by the rate of branching:

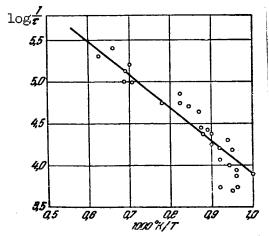
$$\frac{d (H_2O)}{dt} = \frac{d (H)}{dt} = 2k_2 (O_2) (H).$$

The velocity constant of this reaction was determined in [64, 65]. The activation energy lies in the range from 18 to 16 kcal/mole. The results of a calculation of the dependence of the induction period on the temperature are shown by the straight line in Fig. 13 (E = 18 kcal/mole). As we can see from the graph, good correspondence



[commas represent decimal points]

Fig. 12. Comparison of calculation and experiment for the flow velocity behind a shock wave in a mixture of $200 + 0_2$; $p_1 = 0.15$ atm.

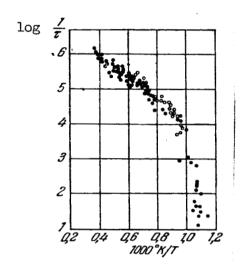


[commas represent decimal points]

Fig. 13. Induction period of the mixture $H_2 + O_2$ as a function of temperature.

Solid line -- calculated.

between the experimental results and those calculated according to the chain theory is observed in the high-temperature region. Analogous data are also obtained in diluted mixtures, where the measurements are less reliable owing to the effect of the diluents [57, 61].



[commas represent decimal points]

Fig. 14. Induction period of hydrogenoxygen and hydrogen-air mixtures as a function of temperature.

-- data taken from [66];
 o -- from [55].

In the low-temperature region, the delays increase more rapidly than we might expect from the calculations. The same conclusion is arrived at in [66] from a comparison of the measurements of a number of investigators. We may cite summarized data on the ignition of a mixture of hydrogen with oxygen, taken from [66], and supplement them with our own results [55] ([66] refers only to the experiments of [59] in the incident shock waves). As can be seen from the graph in Fig. 14, the kinetic curve has a characteristic point of inflection in the region $T \sim 1100^{\circ} K$. In this case the variation in the reaction rate is linked with an approach to the limit owing to the formation of the radical HO_2 and the suppression of branching. There is also a

qualitative change in the nature of the explosion in the gas: whereas far from the limit detonation generally results from the first focus to arise, close to the limit the relatively slow development of a series of ignition centers is always observed. In the region of variation of the rate of the leading reaction, estimating the effective activation energy from the slope of the kinetic curve will be unjustified; hence the value of 90 kcal/mole, obtained for the low-temperature region in [66], should be considered too high.

We should mention that during the induction period (see Fig. 11) the change in the density of the reacting mixture is less than 1%. This shows graphically that the accumulation of active reaction centers occurs with negligible liberation of heat (the thermal effect of the gross reaction is expended on the formation of high nonequilibrium concentrations of hydrogen atoms).

Thus, the data obtained confirm the correctness of the chain theory all the way up to pressures and temperatures of the order of the detonation values. Hence it is natural to expect that kinetic limit phenomena will also involve certain hydrodynamic characteristics of detonation waves (see Section 6.4).

4.2. Determination of the Kinetic Reaction Parameters

Using the results of experiments in which the dependence of the induction period on the temperature is determined, we can find the value of the effective activation energy of the leading reaction from the slope of the curve in the coordinates log 1/T and 1/T. For certain reactions, the path of the experimental curve is close to a straight line, while the value of the activation energy obtained corresponds to the values determined by other methods (for example, the reactions of hydrogen with chlorine, bromine, and iodine [67]). However, in certain cases, for example, in the ignition of a mixture of hydrogen, carbon monoxide, and certain hydrocarbons with oxygen, as we have seen for the example of hydrogen, the dependence proves more complex. Hence the determination of the activation energy for such reactions is more difficult.

If the reaction mechanism is known, the experimental data on the value of the induction period can also be used to determine the pre-exponential factor in Arrhenius' law for the temperature dependence of the rate of the regulating reaction. From the data cited above on the ignition of a mixture of hydrogen and oxygen, for example, the quantity

 $k_2 = 1.8 \cdot 10^{-10}$ cm³/sec, which corresponds to [64] and [65].

Composition of mixture	H +0	20+02	CH, +20	с ₂ 5 +30 ₂	C _T H ₊₁₁₀ + +97% Ar	C ₃ H ₄ ++ + air
Temperature of gas in ignition ex- periments, ^O K	1000 - 1540	1200 - 1400	1050- 1200	1100 - 1300	1500-2400	880-1030
Gas pressure, atm	1+0,2	4-0.5	7 <u>-</u> 0.2	5.8±0.3	2÷20	7 Γ
Effective activation energy for induc- tion period, kcal/mole	17.3±1.5	Varies	27.4+2.5	11.5±2.0	30	19
Bibliography	55	3, 56	2	56	58	54

The table presents experimental values of the activation energies for certain mixtures and indicates the range of thermodynamic parameters of the state of the gas, for which ignition was studied.

4.3. Development of the Detonation Front

As we can see from Fig. 11, after the appearance of a center of ignition, a detonation wave develops in the volume of the adiabatically heated gas mixture. Depending on the temperature and gas pressure, the time of formation of the detonation front varies from several microseconds to a value of the order of 100 μ sec. We should mention that in undiluted, actively reacting mixtures, the detonation wave is formed within the volume of the compressed mixture, independently of the front of the shock wave producing heating of the gas. In diluted mixtures, a gradual acceleration of the reflected shock wave up to the detonation velocity is observed after fusion of the region of gas ignition with its front [58, 61].

When mixtures are ignited under low-temperature conditions, detonation develops in two stages: first, the expanding combustion products adiabatically compress and heat some volume of the unburned mixture adjoining the flame front, and then the compressed gas explodes, and a detonation front is formed, analogously to ignition in shock tubes. In the literature, ignition of this type in shock waves has been termed the transition from slow burning to detonation.

5. TRANSITION FROM DEFLAGRATION TO DETONATION IN GASES

The formation of detonation waves as a result of the acceleration of flames in tubes has been the subject of extensive studies. For the most part, these have been concerned with clarifying the cause of the acceleration of the visible motion of the flame with respect to the walls of the tube [68-75], as well as attempts to construct the most complete gasdynamic scheme of the process [7, 76-83]. Of the greatest interest here are the works of K. I. Shchelkin on the effect of turbulence on the variation of the conditions of flame propagation [68, 69]. Without considering in detail the history of the problem, let us note the sequence of the individual stages of this complex, non-steady-state process. A detailed analysis of the phenomenon has become possible only thanks to the use of shadow methods of investigation, complemented

/538

by pulsed pressure measurements in the gas [3, 4, 26, 83].

5.1. Gasdynamic Scheme of the Development of Detonation in a Tube

On the ignition of a mixture, a laminar flame is formed, the normal propagation velocity of which with respect to the particles of unburned gas is of the order of 1-10 m/sec for different mixtures. If the mixture ignites close to the closed end of the tube, expansion of the combustion products can occur only in the direction of the unburned mixture. As a result, a gas flow is created, similar to the expansion of compressed gas in shock tubes. The expansion of the burned gas leads to the formation of a compression wave in the unburned mixture, in which gas particles are entrained at velocities of several hundred meters per second. The flame is also part of this flow; hence its visible velocity is significantly increased.

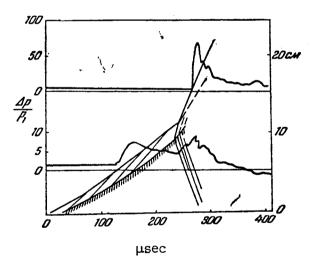


Fig. 15. Schlieren photograph and oscillograms of the pressure during deflagration to detonation transition in a tube.

Mixture of $C_{2}^{H}_{2} + O_{2}^{[83]}$.

<u>/539</u>

It should be noted that the ensuing flow is complex in character, and the state of the burned gas is the resultant after transition first from an unperturbed gas to the compressed state, followed by combustion in the flame front. Hence a simplified analysis of the phenomenon, for example, by replacing the flame with an impermeable piston, moving at the apparent flame velocity [76, 78], is not sufficiently well-founded.

Depending on the actual flow conditions (cross section of tube, roughness of the walls), and on the density and viscosity of the gas in the flow of mixture, turbulence appears, resulting in a change in the flame profile and an increase in its surface. The Reynolds numbers for flows in front of the flame are usually an order of magnitude greater than the critical values, while the length of the sector over which turbulence is appreciable is several dozen tube diameters for smooth walls.

The increase in flame surface corresponds to an increase in compression and a higher flow velocity, which in turn leads to an intensification of the turbulence. Heating of the mixture due to compression also leads to an increase in the normal velocity of the flame. The process develops until the conditions necessary for volume adiabatic spontaneous ignition of the gas are created in the compression wave in front of the flame [55, 83]. The development of turbulence can be accelerated by introducing special baffles in the form of spirals, gratings [67, 75], etc., as well as by the effect on the flame of the compression waves reflected from the opposite closed end of the tube (see Section 5.3).

In Fig. 15 a Schlieren-graph of the process described in a mixture of $C_2H_2 + O_2$ is combined with oscillograms of the pressure, recorded by a piezoelectric pressure gauge at two points along the length of the tube. The volume nature of the explosion of the mixture in front of the flame is evidenced by the shape of the returning compression wave (detonation wave), consisting of several perturbations [83]. It will be noted that the explosion of the gas develops in the compression wave, which has not yet had time to become a shock wave.

5.2. Calculation of the State of the Mixture in Front of the Flame Front

The existing calculation schemes are based on the simultaneous solution of the equations describing the laws of conservation at a double discontinuity -- at the flame front and in the shock wave formed in front of the flame [7, 77]. The simplest calculation scheme for the process is described in [83]. As can be seen from Fig. 15, the pres-

sure drop in the compression-wave front ahead of the flame is small, while the chief increase in the pressure occurs in the region of continuous adiabatic compression. Under such conditions an approximate calculation scheme must suffice: adiabatic wave -- flame -- combustion products at rest with respect to the walls of the tube.

The state of the gas in the compression wave is determined by the flame velocity

$$S = S_n \frac{\Sigma}{\sigma},$$

where S_n is the normal combustion rate, σ is the cross-sectional area of the detonation tube, and Σ is the effective flame surface. For example, for a round tube, replacing the flame surface by a hemisphere,

we have $\frac{\Sigma}{\sigma}$ = 2. If we denote the state of the undisturbed gas by the subscript 0, the gas in front of the flame front by 1, and the burned gas by 2, the state of the unburned mixture at the maximum of the adiabatic compression wave 1, of interest to us, will be determined in the adopted scheme from the following laws of conservation:

$$\varrho_{1}S = \varrho_{2} (S - u)$$

$$\varrho_{1}Su = p_{2} - p_{1},$$

$$u = \frac{2}{\gamma - 1} (c_{1} - c_{0})$$

$$\frac{p_{2}}{\varrho_{2}} = \frac{RT_{2}}{\mu_{2}} \cong const.$$

Here u is the flow velocity, c is the speed of sound, T_2 and μ_2 are the temperature and molecular weight of the combustion products, which do not vary greatly with variation in p_1 . Confining ourselves to the approximation $T_2 = T_c$ (combustion temperature of the mixture), we can find the dependence of u and T_1 on the effective flame velocity S. Fig. 16 shows the results of such approximate calculations for the mixtures

(I) $\text{C}_2\text{H}_2 + \text{O}_2$ [83] and (II) $\text{H}_2 + \text{O}_2$. Thus, the conditions for adiabatic explosion will already have been created for the first mixture at $\text{S/S}_n = 4.5$, but for the second mixture -- not until $\text{S/S}_n = 20-25$. It is characteristic that the predetonation distance for a mixture of hydrogen and oxygen is greater than that for an acetylene-oxygen mixture in exactly the same ratio [4].

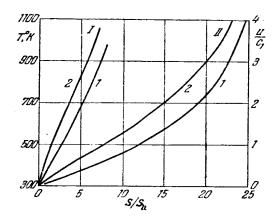


Fig. 16. Approximate calculation of the state of the gas in front of the flame for the mixtures $_2^H_2 + _2^O$ (I) and $_2^H + _2^O$ (II).

1 -- temperature; 2 -- flow velocity of the gas, u/c_1 .

5.3. The Compression Wave and Adiabatic Self-Ignition

The above-described mechanism of the development of detonation on the explosion of an adiabatically heated gas mixture is the most common and is manifested especially distinctly in stoichiometrically active mixtures. In this case the shock front of the wave in front of the flame does not have time to form during the transition; hence the adiabatic scheme of calculation is the most appropriate. Compari-

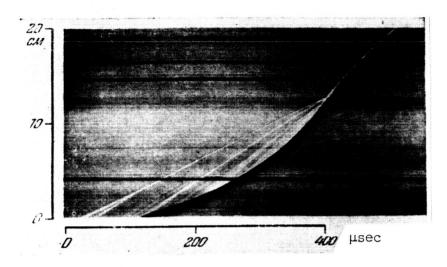


Fig. 17. Development of flame motion in the case of formation of a compression wave-flame complex (Schlieren photograph)

Mixture $C_2H_2 + O_2$; $p_1 = 30 \text{ mm Hg.}$

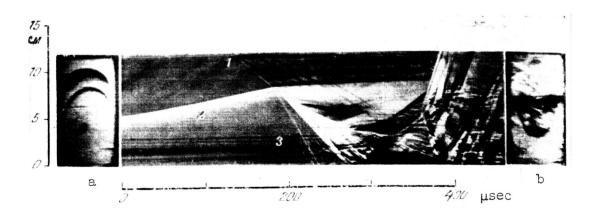


Fig. 18. Schlieren photograph of the refraction of a shock wave at flame front (development).

1 -- front of shock wave; 2 -- flame before collision; 3 -- refracted wave. Frame photographs of the shape of the flame front before and after collision are presented at each side.

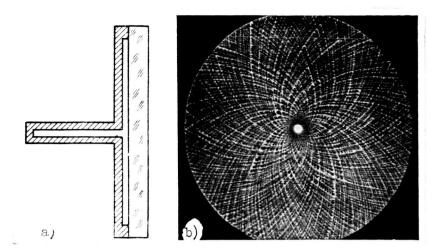


Fig. 19. Scheme of experiment (a) and photograph of luminescence (b) in a radiating detonation wave.

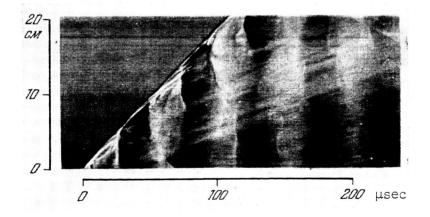


Fig. 20. Development of spinning detonation in a mixture of 2C0 + 0 in a round tube (Schlieren photo-2 graph).

son with the experimental results for a mixture of $^{\circ}_{2}^{\circ}_{2}^{\circ}_{2}^{\circ}$ shows that owing to the influence of local inhomogeneities in the vicinity of the flame and especially close to the walls the gas usually explodes at flame velocities 20-30% lower than the critical values (temperature difference of about 100°C) [83].

In mixtures with a smaller reaction rate and with less heat liberation, the shock front of the compression wave may form during the predetonation stage; in this case, also, however, the maximum pressure and temperature will be reached close to the surface of the flame in accelerated motion. In this case the scheme for calculating the state of the gas is corrected to allow for the appearance of the shock This does not mean that a leading role should be attributed to the formation of a shock wave in front of the flame when detonation occurs. As has already been noted, the detonation wave arises as a result of the explosion of the compressed mixture, and not after fusion of the flame with the shock wave, as occasionally believed [7, 71, 77]. It is interesting that in certain cases the accelerating region of combustion can overtake the front of the compression wave and be propagated together with it for some time as a single complex, without detonation [75]. A development of such a process in a mixture of C₂H₂ + O₂, made by the Schlieren method, is shown in Fig. 17.

5.4. Interaction of Flame and Shock Wave

Investigations in this direction are associated with the special features of the transition from deflagration to detonation in bounded spaces. The compression wave, created by the flame in a tube of finite length, undergoes a series of refractions through the combustion region, producing additional deformation of the flame surface and increasing the normal combustion rate as a result of the increase in temperature (both effects increase the velocity S). The increase in the flame velocity is related to the appearance of additional compression of the gas, which is recorded in the experiments in the form of an intensification of the refracted waves [84-86]. For a relative evaluation of the influence of different factors on the increase in flame velocity, it is advisable to conduct investigations of the interaction of shock waves with the flame in shock tubes [87, 88] when the state behind the shockwave front is given independently and is well known. Fig. 18 shows the development of the collision of the shock wave with the flame in a mixture of C2H2 + O2, made by the Schlieren method. The flow after the first collision of the wave with the flame is in good agreement with the one-dimensional gasdynamic calculation of the collapse of an arbitrary

discontinuity, in which the flame is considered as a contact surface after collision. The chief intensification of the shock wave is observed after its reflection from the end of the tube and secondary refraction across the combustion region (explosion of the gas and the development of detonation behind the reflected wave). The effect of intensification of the reflected wave depends chiefly on the deformation of the flame contour, producing an increase in the surface of combustion. For shock waves of small amplitude, the increase in the normal flame velocity plays a smaller role. This is graphically manifested in a sharp reduction of the intensification of the shock wave after its double refraction through the flame when the site of collision is displaced toward the reflecting wall, the possibility of non-one-dimensional deformations of the flame surface then being practically excluded [88]. The special features of the intensification of the compression waves in the turbulent zone of combustion are discussed in [89].

It is characteristic that in mathematical studies of the problems of the intensification of acoustic and shock waves during their interaction with the flame [85, 86, 90], only the change in the normal combustion rate with the temperature is considered, while the effect of flame deformation is disregarded. At the same time, the role of the instability of the flame front in the processes of transition from deflagration to detonation is quite well known [68, 69]. Thus, in the case of the propagation of a spherical flame [24] or flames in tubes of large cross section, the increase in the flame surface due to the appearance of internal instability, according to L. D. Landau [91], apparently remains the basic cause of the increase in the flame velocity.

6. OSCILLATION OF THE GAS BEHIND THE DETONATION FRONT

The unidimensionality of the flow behind a plane detonation front is usually disturbed due to the appearance of oscillations in the gas. Numerous observations [92-103] give evidence of the fact that the oscillational or "spinning" regime of detonation is the most widespread form of detonation combustion. It has also been revealed that the amplitude and frequency of the oscillations are not only determined by the pressure and chemical composition of the gas, but also depend on the conditions of gas flow behind the detonation front. For example, in the case of detonation in shock tubes, added compression of the burned gas leads to an amplification of the oscillations [26] and to the appearance of secondary pressure waves [95]. In addition, it is well known that the oscillation frequencies can coincide in a number

<u> / 542</u>

of cases with the natural-oscillation frequency of the volume of burned gas. We should emphasize immediately that the source of the oscillations in the gas is the layers adjoining the detonation front, i.e., the reaction zone; hence the cause of the appearance of oscillations should not be sought in the characteristics of the flow of the burned gas. In the case of the "radiation" of oscillations by the detonation front, sometimes the parameters of the radiated waves can only match the frequencies of the oscillations in the burned gas. In such cases the acoustic phenomena are most clearly expressed as a result of the appearance of self-oscillating systems. We shall take up this problem in greater detail in Section 6.3.

6.1. Causes of the Appearance of the Oscillating Combustion Regime

The flow of gas behind the detonation front proves to be hydro-dynamically unstable [104-107]. This instability is related to the existence of a finite chemical reaction zone and an induction period of ignition. As K. I. Shchelkin has shown [104], the boundaries of the appearance of oscillations behind the shock front can be evaluated in the following way.

Steady-state combustion in a detonation wave can be disturbed if the change in the induction period of the mixture, caused by the change in temperature in the disturbances that arise, is of the order of the induction period itself. The range of the pressure variation behind the detonation front can be evaluated by the difference between the pressure of the unburned gas and the Chapman-Jouguet pressure, $p_{sp} - p_{d}$. Hence the criterion of the loss of stability can be expressed in the form

$$\frac{d\tau}{dT} \left(T_1 - T_{sp}\right) \ge \tau \quad \text{or} \quad \frac{E}{RT_{sp}} \left[1 - \left(\frac{p_d}{p_{sp}}\right)^{\frac{\gamma-1}{\gamma}}\right] \ge 1,$$

where E is the effective activation energy for the leading reaction of the induction period. This condition is fulfilled sufficiently well for E > 15-20 kcal/mole, i.e., for most of the mixtures used in laboratory practice. The stability of the detonation front in the presence of small disturbances was investigated in [106]. The structure of the reaction zone in this work differs little from K. I. Shchelkin's

simplified scheme [104]. V. V. Pukhnachev [105] has examined the problem of the stability of the Chapman-Jouguet wave with a one-dimensional, finite reaction zone. Instability was obtained in these investigations for all the cases of detonation of practical interest.

6.2. Properties of Transverse Waves

The chemical reaction zone in a detonation wave is deformed and broken up into a series of disturbances, moving over the surface of the shock front. For a mixture with a high reaction rate, the oscillations that appear have low amplitude and high frequency, while in slowly reacting mixtures and close to the concentration or pressure limits, the amplitude of the oscillations increases significantly, while their frequency drops.

The characteristic dimension determining the oscillation frequency is related to the extent of the chemical reaction zone. The linear dimensions of the vessel in which the detonation wave is propagated influence the frequency and nature of the vibrations only when the average distance between two successive disturbances becomes comparable with the least dimension of the vessel, for example, with the tube diameter [98]. Such a condition is satisfied, for example, by spinning detonation, which will be discussed separately.

The oscillation frequency and the average dimensions of the "cells," into which the detonation front is broken up, depend chiefly on the properties of the mixture. The most convincing evidence of this is the results of experiments with a radiating detonation front [25], when there are no reflecting boundaries in two or three measurements. In the experiment, the scheme of which is shown in Fig. 19, the detonation wave is created in the tube and then emerges into the center of a shallow flat circular channel. The plane of the channel is focused on fixed photographic film. A series of luminous points, the traces of which are outlined on the film of the trajectory in the form of logarithmic spirals

$$r \frac{d\varphi}{dt} = c$$

runs along the detonation front in the transverse direction. The luminous points are the points of intersection of the transverse waves with the detonation front.

The tangential velocity of the disturbances is constant on the average and somewhat exceeds the speed of sound in the burned gas.

The average number of transverse waves per unit length of circumference of the front, with the exception of the vicinity of the channel center, remains constant and depends only on the composition and initial pressure of the mixture. The trace imprints obtained in the reflection of spherically radiating detonation waves from a blackened wall are also evidence of the cellular form of the surface of the detonation front.

Now let us consider the condition of existence of a nondamping detonation with discontinuous ignition behind the front. The combustion of the gas behind the shock front of such a wave is localized chiefly in the region connecting the transverse compression wave with the shock front. Together with the ignition region the transverse wave forms a stable self-oscillating system, in which the thermal energy liberated in the gas is partially used to maintain the transverse wave in accordance with the well-known Rayleigh principle, requiring the liberation of heat in phase with the compression for the excitation of acoustic vibrations in the gas.

If the average velocity of the ignition region is higher than the speed of sound in the burned gas, it will "separate" from the acoustic wave and lose support on the flank. If, on the other hand, the ignition has a lower transverse velocity, the compression wave, having outdistanced the region of ignition, loses its energy source and is damped. Thus, a self-sustaining regime is possible only in cases in which the average velocity of the transverse waves is equal to the speed of sound in the burned gas, taking their mutual collisions into consideration [3, 108].

The relationship between the acoustic phenomena and the special features of ignition behind the detonation wave front is most distinctly manifested in the case of spinning detonation in round tubes. In contrast to waves in flat channels, a self-oscillating system arises, related to the phase wave of the acoustic vibrations. It is interesting to note that a phase wave with a compression antinode close to the wall has a transverse velocity almost twice as great as the speed of sound in the burned gas.

6.3. Acoustic Theory of Spinning Detonation

The transverse velocity of the ignition section in spinning detonation in round tubes is determined experimentally from the angle of inclination to the generator of the spiral track left by the wave on the side surface of a blackened tube. For most mixtures, in which stable rotation of a single transverse wave is observed ("one-headed" detonation), the angle of inclination is close to 45°, i.e., the transverse velocity is close to the detonation speed [92]. An ex-

planation of this fact is given by the acoustic theory of spinning detonation proposed by N. Manson (see [109, 107, 110]). We should mention that the acoustic treatment of spinning detonation in the form presented in [107, 109] indicates only a quantitative relationship between the transverse velocities of the ignition zone and the parameters of the acoustic waves in the burned gas. This coincidence cannot be used as a basis for asserting that the appearance of natural oscillations in the volume of the burned gas is the cause of the appearance of spinning combustion in the detonation wave. Here we have only one case of the appearance of internal instability of combustion behind the shock front under the most favorable conditions. Hence a physical analysis of the causes of the appearance of oscillations and transverse waves in the case of detonation should be related primarily to the characteristics of flow in the zone of the chemical reaction behind the shock front, and not to the processes in the burned gas.

Fig. 20 shows the development of the flow for spinning detonation in a round tube, made by the Schlieren method. Behind the detonation front there exists a transverse wave, extending along the generator of the tube and periodically appearing on the development in the form of light vertical bands. This wave, sometimes called a tail, rotates together with the igniting portion of the front. The existence of such a wave agrees with the solution of the wave equation of the radial and tangential oscillations in the volume of the gas burned in the tube. This solution describes the rotation of the phase pressure antinode of a complex oscillation with a transverse velocity determined from the function

$$v_{periph.} = \lambda_k^{Rc},$$

where λ_k are the eigenvalues of the Bessel functions, R is the radius of the tube, and c is the speed of sound.

For the first harmonic, for example, in a mixture of $200 + 0_2$, $v_{periph.} = 1.84 c = 1780 \text{ m/sec}$. The detonation speed of this mixture D = 1787 m/sec, i.e., it practically coincides with the velocity of the transverse wave. From this we can see that in spinning detonation, stabilization of the ignition is accomplished by a phase compression wave, arising in the burned gas.

If a cylindrical insert is introduced into the detonation tube, the transverse velocity of the phase wave should decrease. The corresponding experiments [111] have shown that the observed decrease in the transverse velocity of the tail fully corresponds to the calculation for an acoustic wave in a coaxial tube.

6.4. Structure of the Spinning-Detonation Front

Careful investigations of the shape of the shock front in a spinning detonation wave in a mixture of 2CO + O₂ led to the diagram shown in Fig. 21 [32, 97, 112, 113]. The lines in the figure are the sites of intersection of the surface of the corresponding shock discontinuity with the surface of the channel of a round tube, the inner surface of the tube having been developed into a plane. The arrows show the direction of the incident flow of gas. The region of intense luminescence is hatched. A study of the structure of the front was performed by photography using the Schlieren method and supplemented by recording the pressure at the tube wall. A photographic method was used, in which the translational and transverse displacements of the ignition area were compensated by the corresponding motion of the photographic film at the same resultant speed [112].

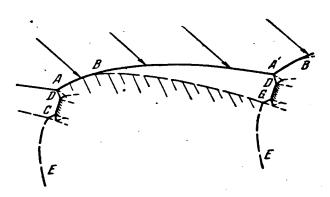


Fig. 21. Structure of the shock front in spinning detonation.

The mixture burns on two sectors of the shock front; in the most curved portion of the basic shock front AB and in the transverse wave DE. This separation of the combustion is related to the kinetic features of the ignition of the given mixture. Gasdynamic calculations have shown that the separation of the ignition front from the basic shock wave T = 1530°K. It must be considered that the sectors of the front with considerable curvature possess a higher temperature, while

/545

in the remaining part of the shock front BG, which forms a sufficiently acute angle with the streamlines of the incident flow of fresh gas, the temperature decreases. As measurements in shock waves have shown [3], in the range $1200^{\circ} < T < 1500^{\circ} K$ the kinetic curve for the mixture $200 + 0_2$ passes from the region of microsecond ignition delays to de-

lays of the order of a millisecond. Thus, doubling of the ignition front is explained by the chemical properties of the given mixture.

The unburned heated mixture, compressed in the past BG of the shock front, ignites in the transverse wave DE, which cuts off the flow. The flanks of the transverse wave form triple shock configurations, bringing the flow of the gas in this wave into agreement with the shock front and the tail [113].

Measurement of the pressure with pulsed piezoelectric pressure gauges with a piezoelement measuring about 1 mm have fully confirmed the flow pattern presented [32]. The maximum pressure measured behind the front of the transverse wave proved to be equal to 160 p, which

is clear evidence of the combustion of the doubly compressed mixture. The measured pressures are significantly higher than the maximum pressures ($\sim 50~p_1$) obtained in calculations introducing a break or

oblique jump in "condensation" in the spinning detonation front [8, 103, 114]. The introduction of triple shock configuration also cannot give a quantity greater than 50 p_1 [114]. The experimental data given

in [32, 113] graphically showed that the Shchelkin-Zel'dovich model [8] is not realized experimentally; anomalously high gas pressures, related to double shock compression of the gas prior to ignition in the transverse wave, exist behind the shock front of the spinning detonation wave.

6.5. State of the Gas in the Detonation Wave Allowing for Transverse Pulsations

Under conditions of substantial nonuniformity of flow, the pulsation components of the velocity, density, and pressure of the gas must be taken into account in the laws of conservation for a shock discontinuity. In this case the final state, described by the Hugoniot equilibrium adiabatic curve, is changed and will differ from the one-dimensional, since a part of the thermal energy liberated is transformed into energy of pulsating motion, which is essentially equivalent to an increase in the effective specific heat of the gas. In addition, pulsating motion creates a supplementary pressure in the medium, the detonation on the whole then being increased. An analysis of the shock

<u> 546</u>

adiabatic curve for detonation assuming pulsating motion behind the front in the form of isotropic turbulence has been performed by D. White [36]. A qualitative picture of the effect of turbulent pulsations on the flow in the wave is given in Fig. 22. Shock transition converts the gas from the initial state 0 to the final state (without reaction) B', the straight line OB' intersecting the equilibrium adiabatic curve of one-dimensional detonation CAD at two points C and D. The intermediate states of the gas in the chemical reaction zone will now lie not on a straight line (of the type of BA in the one-dimensional wave), but on some curve of the type of B'A'D, which passes through point A', where the local Chapman-Jouguet condition is fulfilled (the dotted line shows the shock adiabatic curve on taking into account the loss of part of the thermal effect due to the excitation of turbulence).

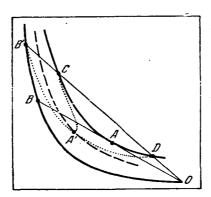


Fig. 22. Shock adiabatic curve for detonation allowing for transverse pulsations behind the front.

After the state A' the pulsating motion is damped and the flow becomes one-dimensional, i.e., the final state approaches the equilibrium adiabatic curve CAD. Here apparently two cases are possible: if the detonation is on the whole self-sustaining, the steady-state reaction zone will culminate in the gas state D on the lower branch of the equilibrium adiabatic curve, while in the case of a small secondary "supercompression" of the detonation, transition to point C on the upper branch will be possible.

The assumption of isotropic turbulence in the combustion zone makes more graphic the picture of the effect of pulsating motion on the gas flow; however, it does not fully correspond to reality. In fact, behind the detonation front there does not exist chaotic turbulent motion -- what arises is a system of acoustic waves with fully definite parameters. Therefore the analysis presented above is mainly of qualitative interest and may be useful in constructing a more complete scheme of the detonation wave.

7. DETONATION IN A STEADY GAS FLOW

Additional data on the conditions of compatibility of gas combustion with an igniting shock wave have been obtained in the study of the detonation created in condensation shocks in continuous flows of a gas mixture. Let us consider the basic lines along which these investigations have been conducted.

7.1. Combustion of Gas Behind a Steady Shock Wave

Combustion of this type has been studied in aerodynamic shock tubes [115-117]. The difficulties of obtaining steady-state detonation in a supersonic gas flow are related to the high Mach numbers for the detonation waves. For stoichiometric mixtures, usually M $_{\rm D}\sim$ 5-7,

while in the case of adiabatic stagnation of the flow behind a shock wave with $M_1 = 3.5-4$ (see Section 4), the gas temperature is already sufficient for rapid autoignition. Thus, the creation of detonation flows is relatively improbable for stoichiometric mixtures, due to the high stagnation temperature and ignition at the walls or during mixing.

If the velocity of the incident flow of the mixture is less than the detonation speed, the detonation wave formed in the case of stagnation at an obstacle separates from the stabilizing body and escapes upward along the flow. Fig. 23 shows successive frames of such a process, obtained under conditions of supersonic flow in the case of stagnation at a step in the shock tube [59].

Steady-state shock waves with ignition of the gas behind the front have been obtained only in diluted hydrogen-air mixtures [115-116]. When hydrogen was added to a supersonic flow of hot air, the condensation shock created at an obstacle changed form, and an extended combustion zone was observed behind the shock front.

7.2. Steady-State Spinning Detonation

By analogy with spinning detonation, B. V. Voytsekhovskiy [118] has devised a scheme of continuous combustion of gas at the detonation speed (Fig. 24). Fresh mixture is continuously supplied to an annular channel in the radial direction through a narrow subcritical gap. The

/547



Fig. 23. Development of detonation behind a condensation shock in a shock tube.

The velocity of the incident flow is less than the detonation speed. Camera speed: 40,000 frames/sec.

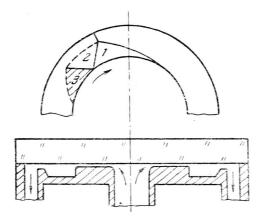


Fig. 24. Scheme of combustion of gas behind a shock wave circulating in an annular channel.

1 -- fresh mixture; 2 -- compressed gas before ignition; 3 -- combustion products.

burned mixture is also exhausted radically. An igniting shock wave, the propagation of which is maintained by the combustion behind its front of the fresh mixture that enters during one rotation of the shock wave, is created in the channel. The velocity of the shock wave in such an experiment proved lower than the detonation speed, since the entry of the fresh mixture and combustion of the gas occur only in a small portion of the shock front. The hatched part of the diagram indicates the combustion region. In the given case, the steady-state hydrodynamic process with combustion of the gas behind the shock wave differs from the Chapman-Jouguet detonation scheme. Such a process is quite stable, and its duration in the experiments described was determined by the reserve of mixture and might amount to several dozen seconds.

7.3. Pulsating Combustion Behind a Shock Wave in a Supersonic Flow

Periodic ignition of the gas behind the front of a shock wave formed at the surface of a body in a supersonic flow has been observed in the ignition of gas mixtures by a flying bullet [119]. In analogous investigations [120], a periodic structure of the flow behind the igniting shock wave was detected. Oscillation of the gas ceased only after a considerable increase in the velocity of the flying body.

A study of this phenomenon, conducted in a shock tube [121], has shown that the periodicity of the ignition is related to the separation of the region of gas combustion from the shock-wave front during its abrupt expansion after ignition. Such a phenomenon of separation of combustion has also been observed in the experiments of [122] in the propagation of detonation in a tube, the cross section of which sharply increased. Separation of fronts was also observed in [123, 124].

Mention should be made of the effectiveness of using shock tubes in experiments with ignition in steady-state supersonic flows. The tube is used in this case as a short-effect wind tunnel. The advantages of this device lie in the fact that the production of high enthalpy in the supersonic flow of mixture created does not involve preliminary heating of the gas to a high temperature before efflux, which makes it possible to conduct the investigation with mixtures prepared in advance, without mixing in the flow.

8. CONCLUSIONS

In concluding this survey of the latest achievements in the field of detonation in gases, we should mention certain problems requiring further elaboration and a search for new solutions. applies primarily to the elucidation of the general conditions of compatibility of chemical and hydrodynamic processes. The universality of the non-one-dimensional mechanism of combustion behind a detonation front remains puzzling. The problem of flow stability in a detonation wave with a finite chemical reaction rate has not been solved sufficiently completely. The mechanism of the relationship between acoustic phenomena in the burned gas and the process of ignition behind the shock front has not yet been clarified. Strictly speaking, in classical detonation this relationship should be absent, since no disturbances should arise behind the Chapman-Jouguet plane. It is possible that losses in the detonation wave play an important role here.

There is no doubt that thermal and diffusion processes must influence the structure of detonation waves with high reaction rates, since large temperature, density, and concentration gradients exist in the detonation front. In this field we as yet have only theoretical estimates [125] and qualitative explanations of certain observations [16].

In the problem of the transition from deflagration to detonation, the general principles of the mechanism of the increase in the flame velocity upon transition to the turbulent regime of gas flow remain unclarified. Should general schemes explaining this process be developed, gasdynamic analysis of the phenomenon and a description of the final, explosive phase of the process should not encounter any serious difficulties.

We should also note the practical importance of the development of various theories of detonative combustion in continuous gas flows. Steady-state combustion behind a sustained shock wave can apparently find application in engines or, for example, in various chemical gas processing operations, in which high-temperature heating, followed by sharp cooling of the mixture during the reaction, is required.

Finally, in a study of the chemical reactions of gases at high temperatures, the application of detonation waves can complement shockwave investigations which have yielded a series of fundamental results in high-temperature kinetics [1, 3, 67]. An important place in these investigations should be set aside for spectroscopic measurements.

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